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Optimal speed of temperature change of a crystal in a pyroelectric X-ray radiation source

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In this work we present the results of the experimental studies of the dependence of the X-ray radiation on the temperature change speed of the lithium tantalate monocrystal in the pyroelectric source of the X-ray radiation. We have found an optimized linear temperature speed change pattern for the pyroelectric crystal designed for generation of the X-ray radiation with enhanced capabilities. The studies that were made open an opportunity to determine the most convenient conditions for generation of X-ray radiation with pyroelectric effect and for development of high-capacity pyroelectric X-ray radiation sources. © *2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).* https://doi.org/10.1063/1.5006486

The effect of X-ray generation combined with pyroelectric monocrystal or ceramics temperature change in vacuum was well studied[1–8] and this effect is considered promising for production of small-size X-ray radiation sources.[9–13] In order to implement the abovementioned effect it is necessary to provide the temperature change for the pyroelectric material (the typical size of the object is around 1 cm) of several dozen of °C while the pressure of the environment is expected to be of the order of several mTorr. These conditions guarantee generation of the electrical field of the order of 10^5 V/m in the space between the ungrounded surface of the pyroelectric material and the grounded target. The electrical field is sufficient for ionization of the molecules of the residual gas and the secondary (field) electron emission from the pyroelectric surface which allows to observe X-ray radiation with the energy of up to 300 keV.[14] The mentioned features open new possibilities to develop portable pyroelectric X-ray sources for different applications.[15]

The influence of various regimes of temperature changes of the pyroelectric crystal on the effect of X-ray radiation has not been studied systematically, whereas the temperature change regime determines the electric current magnitude which is generated through the pyroelectric effect as well as the electric field intensity. In most of available papers dedicated to this topic the temperature of a crystal varies when the thermoelectric cooler (TEC) current is constant (except for paper[16] where the fixed stabilized time temperature gradient was used). The TEC is used to drive the temperature of pyroelectric crystals up or down. Based on the speed of temperature change $\frac{\Delta T}{\Delta t}$, the area of working surface of the crystal "A" and the pyroelectric coefficient γ we can estimate the electric current which is generated during the pyroelectric effect:

$$i = \gamma A \frac{\Delta T}{\Delta t}. \quad (1)$$

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According to (1) the increase of the temperature speed change should increase the electric current which is generated due to the pyroelectric effect which should also increase the output of the X-ray radiation.

We present the results of the experimental studies of the dependence of the X-ray radiation on the temperature change speed of the lithium tantalate monocrystal in the pyroelectric source of the X-ray radiation.

The main component of the pyroelectric source is comprised of a lithium tantalate monocrystal ($LiTaO_3$). We use is a cylinder diameter of 20 mm and 10 mm in height for the sample model. The Z-axis of the crystal is parallel to the axis of the cylinder. The change of the crystal temperature was performed by TEC. The target was made of a copper plate with the following dimensions: 45*45*0.5 mm. Heat removal from the TEC was carried out through the radiator (heat sink) made of duraluminium through the vacuum chamber into the environment. Heat transfer from the TEC to the crystal was done through the heat conductor made of duraluminium (with diameter of 20 mm and height of 4 mm). Some elements of pyroelectric source: the crystal, the heat conductor, the TEC, and the radiator were mounted to each other with heat- and electro-conducting glue. The measurement of X-ray spectra was done through an X-ray spectrometer which consisted of a semi-conductor X-ray radiation detector Amptek Cd-Te XR-100T. The latter was equipped with a 25 mm^2 crystal (collimated to 17 mm^2) and PX5 digital pulse processor which was connected to the computer. The detector was mounted at 330 mm from the axis that went through the crystal and the target. The specific linear change of temperature was controlled by a special device – a heat controller which supplied the TEC using the feedback provided by the thermocouple. The scheme of the experiment is described in Figure 1.

If we used the thermocouple on the surface or within the volume of the crystal it disrupted the configuration of the electric field and deteriorated the generation of the X-ray radiation which would also make the fair evaluation of the processes we studied impossible. Because of that the thermocouple was mounted to the heat conductor close to the lower surface of the crystal. The change of the crystal temperature can also be considered linear with a stipulation that the real speed of the crystal temperature was slightly lower than that measured during the experiment due to the weak heat conductivity of the crystal itself.

All the measurements were made under the residual gas pressure of 1+/-0.3 mTorr. At each measurement the temperature of the crystal was changed by the same magnitude of 15 °C in order to provide the same amount of charge which was generated by the pyroelectric effect, hence the time of measurement was different for each separate experiment. The time t for each spectrum depended on the speed of temperature change $\frac{\partial T}{\partial t}$ for the crystal in the experiment.

$$t = \frac{15}{\frac{\partial T}{\partial t}}. \tag{2}$$

The initial temperature at the start of the heating process was in the range of 28.2±0.8 °C, and the one before the cooling was 30.6±0.9 °C. The measurements were conducted for ten different values

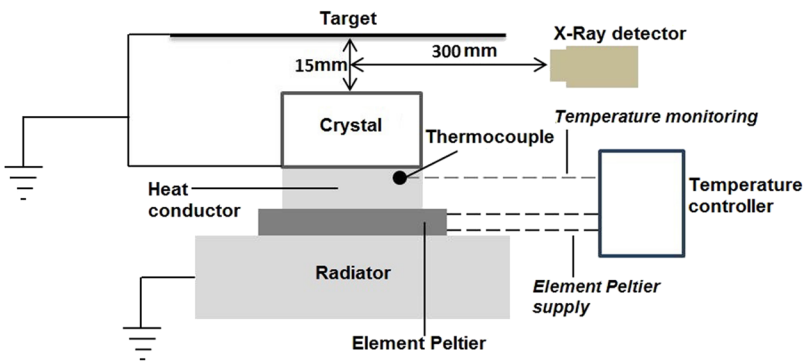

FIG. 1. Experimental scheme.

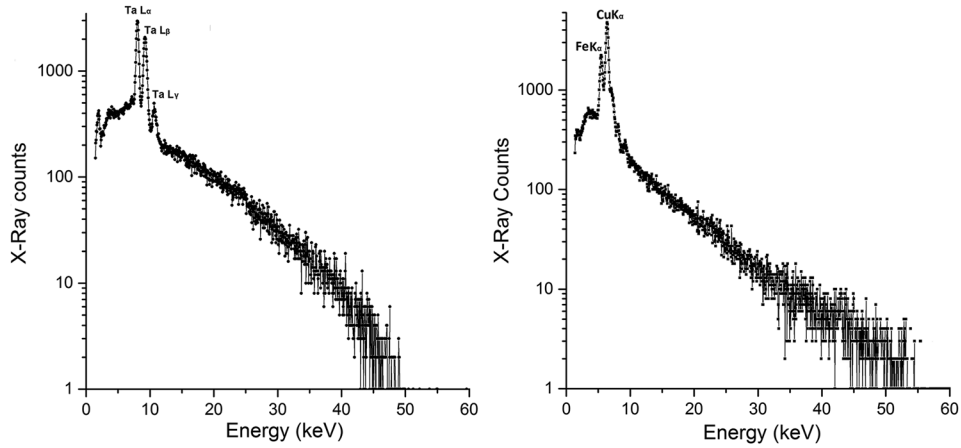

FIG. 2. Typical X-ray spectra at heating (on the left side) and cooling (on the right side) of pyroelectric crystal.

of the crystal temperature speed changes in the ranges from 2°C to 22 °C per minute during heating and from 0.5°C to 10°C per minute during cooling. When the measurements were done for different speeds of temperature changes the vacuum chamber was filled with atmospheric air to prevent the former measurement to affect the latter. The difference in the temperature diapasons for different polarities is explained by the technical specifications of the working TEC mechanism. Heating was used when the free surface of the crystal had positive polarity, whereas cooling corresponded to the negative polarity.

Below in Figure 2 we have present the typical spectra of the X-ray radiation for both heating and cooling of the pyroelectric crystal whereas Figure 3 shows the dependency of the overall number of quanta of the X-ray radiation recorded for a particular speed of temperature change of the pyroelectric crystal.

The output of the X-ray radiation generally grows monotonically when the speed of change of the temperature of the crystal is up to 7-8 °C per minute. At the 7-8 °C per minute we suddenly observe a sharp maximum of the X-ray radiation output (up to 170000 events which were registered by the detector during the time of measurement) for both positive and negative polarities of the crystal surface. For other speeds of temperature change we observe $5 \cdot 10^4$ events per heating-cooling cycle, except for the following points: 16 °C per minute for the positive polarity and 0.5°C and 4°C per minute for the negative charge. The observed yield of X-ray radiation is typical for pyroelectric sources.[15]

Below in Figure 4 there we show the dependencies of the endpoint energy which were recorded during a particular measurement of the temperature speed change of the pyroelectric crystal.

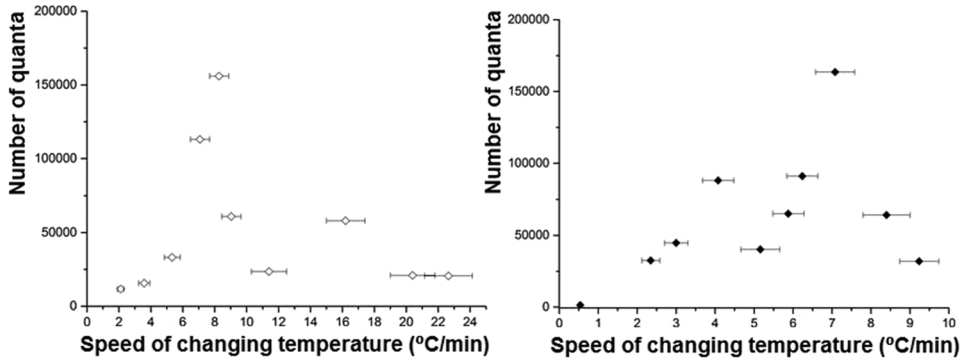

FIG. 3. The dependence of the X-ray output (number of quanta) on the speed of temperature change of the pyroelectric crystal. The positive polarity of charge on surface of crystal is on the left side and the negative one is on the right.

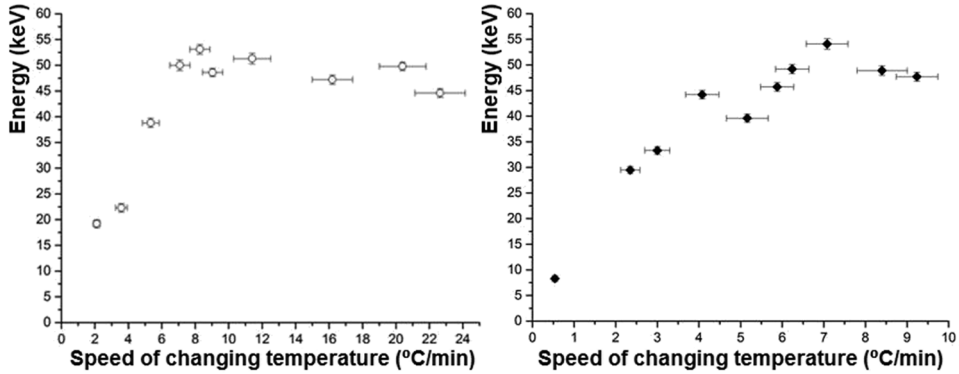

FIG. 4. The dependence of the endpoint energy on the speed of temperature change of the pyroelectric crystal. The positive polarity of charge on the crystal surface is on the left side, negative is on the right side.

The endpoint energy initially grows monotonically simultaneously with the increase of the speed of temperature change up to 5 °C per minute and then reaches the plateau and does not go lower than 45 keV starting from 6 °C per minute for both cases of polarity. At the same time the relative maxima of the endpoint energy fall in the same area of the temperature change speeds which are 7-8 °C per minute. In case of negative polarity charge the dependence of the endpoint energy appears to have the same shape as the dependence of the X-ray radiation output. That way both the endpoint energy and the X-ray radiation output depend on the temperature change speed of the pyroelectric crystal and at the same time there exists an area of speeds where the parameters of the generated X-ray radiation reach their maximum values.

Thus, our research demonstrates that there exists an optimized pattern for the speed of temperature changes for the pyroelectric crystal in the pyroelectric X-ray source and with this optimized speed the output of the X-ray output is 3 times greater compared to an average X-ray radiation output. When speeds which are less than optimal are applied, the maximum energy and the output of the X-ray radiation increase with the increase in the speed of temperature change (monotonically on average). Going over this speed does not provide either the increase in X-ray radiation output or the maximum energy of the X-ray radiation because the dependence of the endpoint energy reaches a plateau and the radiation output gets smaller. In our case the optimal speed is 7-8 °C per minute.

Thus, when linear temperature change of a pyroelectric crystal is applied, the speed of the temperature change is the key factor which affects the generation of the X-ray radiation taking into consideration the pyroelectric factor. We found out experimentally that there exists a certain area of the crystal temperature change when the parameters of the generated radiation reach their maximum values. It is still an open question which parameters determine the area of location of the optimal speed and what is the physical mechanism of its appearance. In the future it is necessary to figure out experimentally the dependencies of the generated X-ray radiation on the speed of the temperature change for the test specimens with various geometrical parameters, different mechanical conditions of the specimen's surfaces, and for various pressure levels of the residual gas.

From one point of view, the studies which were made open an opportunity to determine the most convenient conditions for generation of the X-ray radiation with the pyroelectric effect and for development of the high-capacity pyroelectric X-ray radiation sources. On the other side, the obtained results open new areas of pyroelectric effect in monocrystals and require a more thorough physical explanation and understanding.

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